

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of

Tsukasa MARUYAMA

Group Art Unit: 1728

Serial No. 10/585,121

Examiner: CHAN, HENG M

Filed: June 30, 2006

For: Electrolyte For Photovoltaic Device As well As
Photovoltaic Device And Dye-Sensitized Solar Cell Including
That Electrolyte

DECLARATION UNDER RULE 132

Honorable Commissioner of Patents and Trademarks

Washington, D. C. 20231

sir:

I, Tsukasa MARUYAMA, a citizen of Japan, residing at
c/o THE YOKOHAMA RUBBER CO., LTD. Hiratsuka Factory, 2-1,
Oiwake, Hiratsuka-shi, Kanagawa, Japan, respectively,
sincerely and solemnly declare:

That I am by profession a research chemist and
that I graduated from Tokyo Institute Technology, Faculty

of Engineering, Department of Polymer Chemistry on March, 1988 and finished Master Course of Tokyo Institute of Technology, Department of Electronic Chemistry, on March, 1990 and have a PhD degree received from Tokyo Institute of Technology on May, 1998.

That, since August, 1998, I have been employed by THE YOKOHAMA RUBBER CO., LTD. and have been engaged in research mainly into the development of rubber additives, electrolytes for photovoltaic devices, and batteries and now a general manager of Maruyama Laboratory at Research and Development Center of THE YOKOHAMA RUBBER CO., LTD.:

THAT I am one of the inventors of the invention as disclosed and claimed in the above-mentioned application (i.e., the present application) and the Office Action mailed July 18, 2011.

In order to show the superiority of the present invention over the Prior Arts, the following Experiments were carried out under my direction and supervision.

EXPERIMENT I (Present Invention) and Comparative

Experiments I - III

Preparation of Electrolytes A - D

(1) Preparation of Clay Gel Electrolyte A

0.1 mol/liter of lithium iodide, 0.15 mol/liter of iodine and 0.5 mol/liter of 4-tert-butylpyridine were dissolved in an ionic liquid 1-methyl-3-propylimidazolium iodide to prepare an electrolyte. The organically modified layered clay mineral Lucentite SPN was added to the electrolyte obtained in an amount of 7.5 wt%, followed by mixing by a mortar to obtain a clay-like substance "Clay Gel Electrolyte A" composed of the ionic liquid and organically modified layered clay mineral.

(2) Preparation of Electrolyte B (Comparative)

An "Electrolyte B" was obtained in the same manner as in the above preparation of Clay Gel Electrolyte A, except that carbon black (i.e., SEAST KH available from Tokai Carbon Co., Ltd.) was used instead of the organically modified layered clay mineral Lucentite SPN.

(3) Preparation of Electrolyte C (Comparative)

An "Electrolyte C" was obtained in the same manner as in the above preparation of Clay Gel Electrolyte A, except that talc (i.e., general purpose talc SSS available from Nippon Talc Co., Ltd.) was used, instead of the organically modified layered clay mineral Lucentite SPN.

(4) Preparation of Electrolyte D (Comparative)

An "Electrolyte D" was obtained in the same manner as

in the above preparation of Clay Gel Electrolyte A, except that calcium carbonate (i.e., Hakuenka A available from Shiraishi Calcium Kaisha, Ltd.) was used instead of the organically modified layered clay mineral Lucentite SPN.

Fabrication of Cell

Fabrication of Photoelectrode

A transparent conductive glass substrate (made by Nippon Sheet Glass, conducting layer made of fluorine-doped tin oxide, sheet resistance $8\Omega/\square$) was coated on its surface with a paste prepared by mixing titanium dioxide powder (AEROXIDE TiO₂ P25, average particle size 25 nm, made by Nippon Aerosil), acetyl acetone, distilled water and a surfactant (Triton X100, made by Acros Organics) by a mortar, dried, then sintered at 460°C for 45 minutes, whereby the transparent conductive glass substrate was formed with a mesoporous titanium oxide thin film. The glass substrate having this mesoporous titanium oxide thin film was dipped in an ethanol solution (concentration of 3×10^{-4} mol/liter) of ruthenium complex dye (i.e., cis-di(thiocyanate-N,N'-bis(2,2'-bipyridyl-4,4'-dicarboxylic acid) ruthenium (II) complex, Ruthenium 535, made by Solaronix) for 12 hours. The result was washed with

acetonitrile, then dried in a dark location in a flow of nitrogen to impregnate the mesoporous titanium oxide thin film with a sensitizing dye. This was used as a photoelectrode.

Fabrication of Platinum Counter Electrode

A transparent glass substrate (made by Nippon Sheet Glass, conductive surface made of fluorine-doped tin oxide, sheet resistance $8\Omega/\square$) was formed on its surface with an approximately 100 nm platinum thin film by sputtering. This electrode was used as a counter electrode.

Experiment I (Present Invention)

The fabricated photoelectrode was coated with the Clay Gel Electrolyte A prepared above was superposed on the platinum counter electrode, then was fastened by clips to obtain the cell of Experiment I (using the Clay Gel Electrolyte A).

Experiment II (Comparative)

A cell was prepared in the same way as in Experiment I, except for using the above Electrolyte B instead of the Clay Gel Electrolyte A.

Experiment III (Comparative)

A cell was prepared in the same way as in Experiment I, except for using the above Electrolyte C instead of the

Clay Gel Electrolyte A.

Experiment IV (Comparative)

A cell was prepared in the same way as in Experiment I, except for using the above Electrolyte D instead of the Clay Gel Electrolyte A.

Evaluation of Cells

The cells of Experiments I - IV, were irradiated from the photoelectrode side with AM1.5 pseudo sunlight at a light intensity of 100 mW/cm^2 using a solar simulator as a light source. A current/voltage measurement apparatus (Digital SourceMeter 2400 made by Keithly Instruments) was used to find the short-circuit photocurrent, open-circuit photovoltage, fill factor, and conversion efficiency. The results of the cells are shown in Table I.

Table I

Experiment No.	Electrolyte	Current-voltage characteristic			
		Short-circuit photocurrent density (mA/cm^2)	Open-circuit photovoltage (mV)	Fill factor	Conversion efficiency (%)
I	Electrolyte A	8.6	686	0.64	3.8
II	Electrolyte B	6.5	623	0.52	2.1
III	Electrolyte C	6.2	623	0.56	2.2
IV	Electrolyte D	6.1	612	0.51	1.9

As is clear from the results shown in Table I, when the clay gel "Electrolyte A" was used, the current-voltage characteristic is higher, when compared with those of the comparative "Electrolyte B" (carbon black), "Electrolyte C" (talc) and "Electrolyte D" (calcium carbonate).

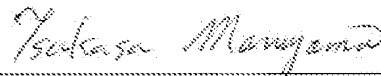
CONCLUSION

From the test results shown in Table I, and based on my own experience and knowledge, I conclude that the use of the clay gel electrolyte is superior to the use of the carbon black, talc and calcium carbonate, instead of the clay gel, because the cell of the above Experimental I using the clay gel shows higher current-voltage characteristic than the cells of Experimental II-IV using the carbon black, talc and calcium carbonate, respectively.

Consequently, I believe that the present invention is by no means obvious from the Prior Arts.

I, the undersigned declarant, declare further that all Statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and; further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18, of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 30th day of November, 2011

A handwritten signature in cursive script, reading "Tsukasa Maruyama", is written over a horizontal line.

Tsukasa MARUYAMA